



## Spatial patterns and sources of atmospheric nitrogen deposition in the Greater Yellowstone Ecosystem, Wyoming determined from lichens

Abigail S. Hoffman and David G. Williams\*

Department of Botany, University of Wyoming, Laramie, WY

\*Author for correspondence: dgw@uwo.edu

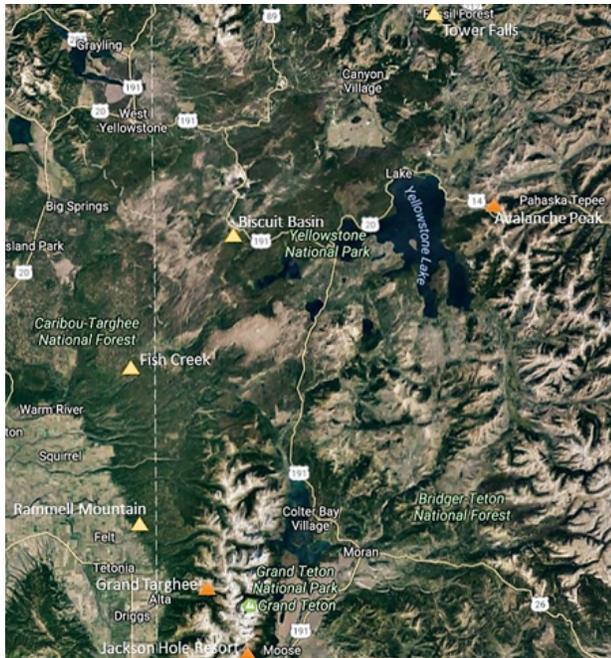
**Abstract** Increased anthropogenic nitrogen (N) deposition can lead to N saturation of ecosystems, altering water quality, biogeochemical cycling and biodiversity. Although some N deposition ( $N_{dep}$ ) is natural, there has been an increase of  $N_{dep}$  in the Greater Yellowstone Ecosystem (GYE), largely due to local and regional intensification of agricultural activity, which releases ammonia ( $NH_x$ ), and transportation and industrial processes, which release nitrogen oxides ( $NO_x$ ). The climate, topography, and sources of  $N_{dep}$  in the region likely create heterogeneous patterns of  $N_{dep}$  in the GYE, where nutrient-limited alpine ecosystems are especially susceptible to  $N_{dep}$ . Epiphytic lichens obtain their nutrients from the air and record local scale patterns of  $N_{dep}$ . We collected 162 lichen samples (*Usnea lapponica* and *Letharia vulpina*) and analyzed them for %N and  $\delta^{15}N$  at 15 sites in the GYE to understand patterns and sources of  $N_{dep}$  in the GYE at small spatial scales. We found that lichen %N was higher closer to the Snake River Plains and at higher elevations, which indicates higher deposition at those sites. This is likely because N is more likely to be deposited closer to major sources and because N is often deposited in precipitation so deposition patterns follow precipitation patterns. Additionally, the mean  $\delta^{15}N$  value was  $-11.8 \pm 3.2\%$ , which suggests an agricultural source of  $N_{dep}$ , but  $\delta^{15}N$  values increased with higher %N, which indicated sites with high deposition were receiving more N from combustion sources. However, the large amount of variation in lichens collected at a single site suggest that future work needs to address how microhabitat factors influence lichen N incorporation.

### Introduction

Perturbation of the nitrogen (N) cycle by human activities affects water quality, ecosystem functioning, biodiversity and human health. Increased atmospheric nitrogen deposition ( $N_{dep}$ ) resulting from human activities is the third most important driver of biodiversity loss globally (Sala et al., 2000). Although some  $N_{dep}$  is natural, there has been an increase in  $N_{dep}$ , largely due to intensification of agriculture activity, where ammonia ( $NH_x$ ) volatilizes off of fertilizers and animal waste, and transportation and industrial processes, which release nitrogen oxides ( $NO_x$ ), through high temperature combustion. This is of particular con-

cern in the Western US where alpine ecosystems are N limited due to poor soil. Increased  $N_{dep}$  in these environments drives changes in plant communities through altered competitive relationships, enhanced leaching of cations, and acidified soils (Bobbink et al., 2010). Air quality in Class I Wilderness areas, including 49 National Parks and Monuments, is protected under Section 162(a) of the federal Clean Air Act. It is important therefore to understand sources of  $N_{dep}$  to mitigate ecosystem alterations and comply with federal law.

In the Western U.S., several studies have documented increased  $N_{dep}$  (Baron et al., 2000, 2013; Burns, 2004). In Rocky Mountain National Park



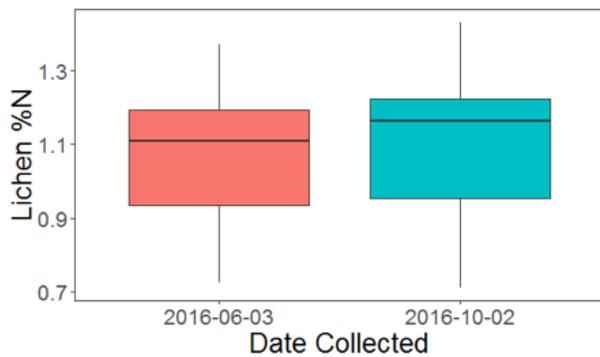
**Figure 1.** Lichen collection sites in the Greater Yellowstone Ecosystem. Orange points indicate elevational transects, which include four collection sites, and yellow points indicate sites that are part of the latitudinal transect.

(RMNP),  $N_{dep}$  has been measured at  $2\text{--}8\text{ kg N ha}^{-1}\text{ yr}^{-1}$ , and National Atmospheric Deposition Program (NADP, <http://nadp.slh.wisc.edu/>) network data shows that  $N_{dep}$  has been increasing since the 1980s (Burns, 2003, 2004). In the Rocky Mountains, in high elevation areas with rocky soils and steep slopes, critical loads of  $N_{dep}$ , where ecosystems are adversely impacted occur at less than  $1.5\text{ kg N ha}^{-1}\text{ year}^{-1}$  (Nanus et al., 2012). Additionally, source modeling suggested that, on average, RMNP receives  $4.0\text{ kg N ha}^{-1}\text{ year}^{-1}$ , while Grand Teton National Park receives  $3.3\text{ kg N ha}^{-1}\text{ year}^{-1}$  (Lee et al., 2016).

Several air pollution monitoring efforts across the U.S. have found significant variation in  $N_{dep}$  over fine spatial scales, however the drivers of fine scale  $N_{dep}$  are not well understood. In the Catskill Mountains, atmospheric deposition varied across the landscape, possibly due to differences in wind and precipitation patterns created by topography (Weathers et al., 2000). Recently, studies in RMNP found that east of the continental divide there are higher levels of

$N_{dep}$ , likely due to precipitation patterns tied to elevation change and the proximity to N emissions from urban areas to the east (Beem et al., 2010; Clow et al., 2015). In addition, vegetation cover may play a role in the amount of N that reaches soils and water because some plants take up atmospheric N through their leaves, while other plants leach N (De Schrijver et al., 2007). Finally, Weathers et al. (2006) created an empirical model using elevation and vegetation data from Acadia and Great Smoky Mountains National Park, to explain  $N_{dep}$  variation over fine scales and found four to six-fold differences in  $N_{dep}$  within each park. These studies suggest that  $N_{dep}$  is heterogeneous within fine spatial scales because it is influenced by many factors, including emission sources, local and regional atmospheric transport, elevation, topography, precipitation, and vegetation type.

Many recent air quality monitoring studies have focused on understanding global and regional patterns of  $N_{dep}$ . There are several national monitoring networks, most notably, Clean Air Status and Trends Network (CASTNET, <https://www.epa.gov/castnet>) and NADP; however, these sites are sparsely located (300 sites across the U.S.) and mostly at lower elevations so they are insufficient to capture  $N_{dep}$  patterns on fine spatial scales due to variation in topography and elevation (Baumgardner et al., 2002). Current atmospheric modeling efforts, such as GEOS-CHEM, a global chemical transport model, and BioEarth, a regional Earth System model, use regional scale atmospheric processes, such as precipitation, air mass transport, and atmospheric chemistry, as well as land feedbacks such as albedo, energy fluxes and biogeochemical cycling (Adam et al., 2015). While these models work well on regional scales, they do not explain fine scale variation in  $N_{dep}$ , which more directly impacts ecosystem functioning and variation in plant communities across the landscape. It is important to study  $N_{dep}$  at small spatial scales in the GYE because  $N_{dep}$  patterns vary greatly at small spatial scales within and between ecosystems due to differences in topography, climate and weather patterns. Understanding  $N_{dep}$  variation across the landscape can help to identify areas that are a priority for conservation due to unique topographical features that protect them from

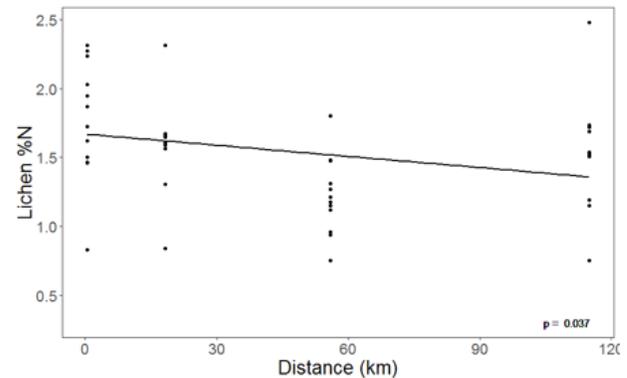


**Figure 2.** Lichen thalli %N for lichens collected in Granite Canyon, Grand Teton National Park on June 3, 2016 (red) and October 2, 2016 (blue).

elevated  $N_{dep}$  and will allow park managers to establish critical load thresholds and  $N_{dep}$  reduction plans.

Several studies have used epiphytic lichens as bioindicators to classify the degree of pollution in an area because lichens obtain their nutrients from the atmosphere so their tissues reflect atmospheric conditions. In the Columbia River Gorge in the Pacific Northwest, areas that had higher N air pollution also had higher N content in lichens, but this relationship was not well developed (Fenn et al., 2007). Additionally, along an urban-rural gradient in England, lichen N concentrations were higher and C/N ratios were lower closer to urban areas with high N emissions (Power and Collins, 2010). A study in the Northern Rocky Mountains compared Community Multiscale Air Quality (CMAQ) modeled  $N_{dep}$  projections to N concentrations in lichens and found some correlation between the two, but suggested that the modeled  $N_{dep}$  values are not accurate on a small scale because the model does not account for local emission sources (McMurray et al., 2015). Analysis of lichen tissues could lead to more precise understanding of variation in  $N_{dep}$  at finer scales than currently possible with air monitoring networks.

Furthermore, lichen tissues may be used to understand sources of  $N_{dep}$ . Several studies have used N isotopes to discern sources of atmospheric N because N fractionates based on how it was combusted (Elliott et al., 2007, 2009). Typically,  $N_{dep}$  volatilized from agricultural processes has a lower  $\delta^{15}N$  value



**Figure 3.** Lichen thalli %N collected at increasing distances from the Snake River Plains, Idaho, which are a major source of  $NH_4^+$ . Lichen %N decreases with distance from the Snake River Plains ( $p = 0.037$ ).

ranging from  $-14$  to  $-8\text{‰}$  in lichens (Boltersdorf and Werner, 2014), whereas  $N_{dep}$  from combustion has a higher  $\delta^{15}N$  value ranging from  $-10$  to  $+20\text{‰}$  in precipitation (Zhang et al., 2008).

In the Greater Yellowstone Ecosystem (GYE),  $N_{dep}$  is increasing, likely due to increasing agricultural activity to the west in the Snake River Plains, oil and gas extraction to the southeast, and increased automobile emissions with local tourism. The Grand Teton Reactive Nitrogen Deposition Study found the values of  $N_{dep}$  in and around the park ranged from  $0.6$ – $0.9$   $kg\ N\ ha^{-1}$  over the 2-month study period (Benedict et al., 2013). Additionally, higher rates of  $N_{dep}$  were observed to the west of the park and lower rates were observed in the eastern part of the park (Benedict et al., 2013). Another study assessing the impact of drilling near Pinedale, WY suggests that drilling elevates  $N_{dep}$  within about 40km of the wells, after which,  $N_{dep}$  drops back to background levels (McMurray et al., 2013). The study also suggests there is a strong relationship between lichen %N and  $N_{dep}$ , but sample sizes are relatively small (McMurray et al., 2013). It is important to understand patterns of fine scale  $N_{dep}$  in such Class 1 Wilderness areas to understand threats to natural resources and accurately predict future impacts. This report presents preliminary data on spatial patterns of  $N_{dep}$  in the GYE as recorded by lichen thalli (vegetative tissue).

site	n	%N	sd	$\delta^{15}\text{N}$ (‰)	sd	elevation (m)
AP1	10	1.51	0.36	-13.17	1.57	2536
AP2	9	1.18	0.15	-15.61	1.52	2612
AP3	6	1.32	0.31	-13.05	1.56	2874
GT1	9	1.17	0.32	-14.46	1.40	2307
GT2	21	1.39	0.31	-11.61	1.54	2450
GT3	12	1.56	0.24	-10.57	1.85	2684
GT4	8	1.55	0.33	-10.65	2.27	2889
JHMR1	19	1.08	0.22	-12.82	1.66	1966
JHMR2	3	1.74	0.15	-12.34	0.82	2109
JHMR3	10	1.73	0.47	-6.50	5.32	2536
JHMR4	6	1.41	0.27	-12.03	2.56	3030
Rammell Mtn	12	1.77	0.43	-11.57	0.96	2092
Fish Creek	7	1.51	0.30	-8.99	4.18	2046
Biscuit Basin	12	1.22	0.28	-14.11	1.67	2332
Tower Fall	11	1.50	0.45	-11.85	0.90	1944

**Table 1.** Means and standard deviations of %N and  $\delta^{15}\text{N}$  for lichen collections at each site in the Greater Yellowstone Ecosystem.

## Methods

### Study sites

We sampled two species of lichen (*Usnea lapponica* and *Letharia vulpina*) at 15 sites in Grand Teton National Park (GTNP), Yellowstone National Park (YNP), Caribou Targhee National Forest (CTNF), and Bridger Teton National Forest (BTNF) from June to October 2016. These sites included elevation transects near Jackson Hole Mountain Resort (JHMR), Grand Targhee Ski Resort (GT) and Avalanche Peak (AP), to understand how elevation impacts N deposition, as well as a latitudinal transect beginning near Rammell Mountain in the southwestern GYE and extending to Tower Falls in the northeastern GYE (Figure 1).

### Collections and analysis

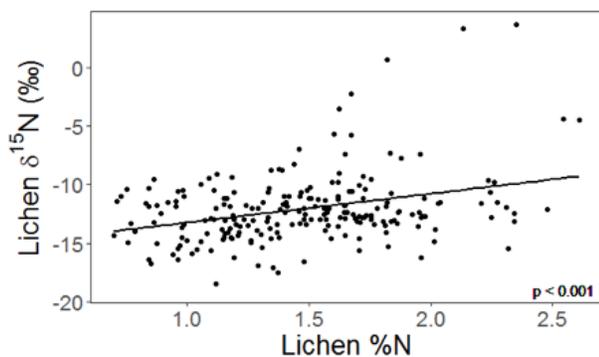
At each site, where possible, we collected 10 samples each of *U. lapponica* and *L. vulpina*, from evergreen trees. Collections were made under permits GRTE-2016-SCI-0024 (Study # GRTE-00388) and YELL-2016-SCI-07010 (Study # YELL-7010). Each

lichen sample was oven dried at 60°C, homogenized with a berry ball machine and analyzed for %N and delta  $\delta^{15}\text{N}$  on a Delta V isotope ratio mass spectrometer.

## Preliminary Results and Discussion

We collected 71 *U. lapponica* and 91 *L. vulpina* at 15 sites across the GYE. The mean %N in lichen thalli for *U. lapponica* was  $1.51 \pm 0.43\%$ , while the mean  $\delta^{15}\text{N}$  value was  $-12.2 \pm 3.4\text{‰}$ . For *L. vulpina*, the mean lichen thalli %N was  $1.33 \pm 0.33\%$  and the mean  $\delta^{15}\text{N}$  value was  $-11.6 \pm 3.0\text{‰}$ .

The %N of lichen thalli collected in Granite Canyon did not differ between June and October samples (Figure 2). The lack of seasonal change in %N allowed us to compare lichen samples collected from different sites at different times during the summer. Typically, lichen grow most during the winter and early spring when temperatures are wet and cool (Caldiz, 2004), so the majority of tissue is likely added during that time, with relatively little tissue growth over the course of the hot, dry summers, such that  $N_{dep}$



**Figure 4.** Lichen thalli %N and  $\delta^{15}\text{N}$ , collected at 15 sites across the Greater Yellowstone Ecosystem.  $\delta^{15}\text{N}$  increases with %N ( $p < 0.001$ ).

reflects the site rather than the time of year.

Along the latitudinal transect, %N in lichen thalli decreased with distance from the Snake River Plains (Figure 3). The mean %N in lichen at Rammell Mountain, Fish Creek, Biscuit Basin and Tower Falls were  $1.77 \pm 0.43\%$ ,  $1.51 \pm 0.3\%$ ,  $1.22 \pm 0.28\%$ , and  $1.50 \pm 0.45\%$ , respectively (Table 1). This indicates that main sources of  $N_{dep}$  in the GYE are to the west, because deposition declines across the GYE.

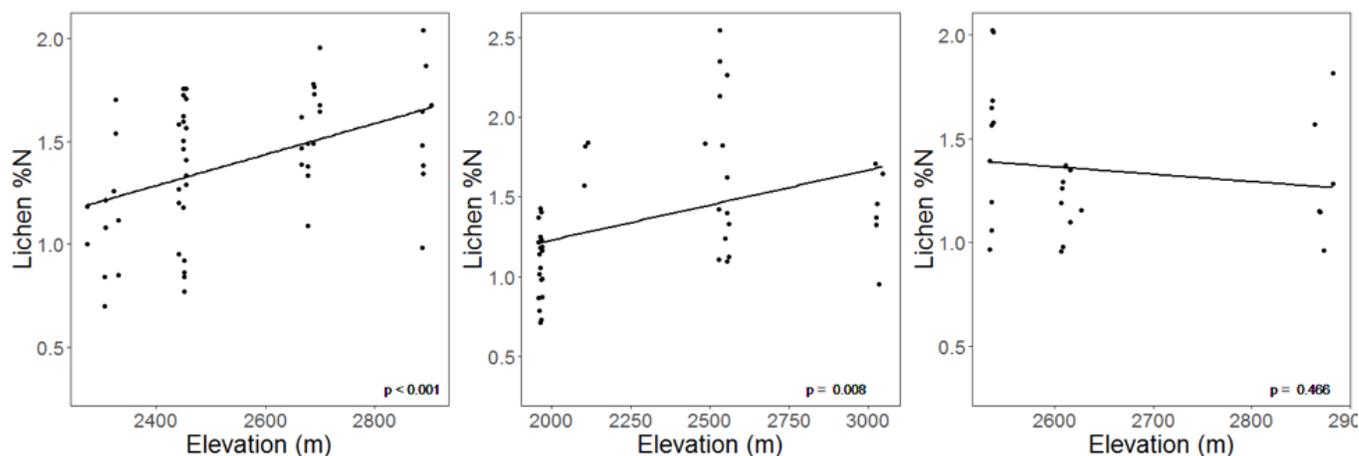
$\delta^{15}\text{N}$  values were generally low, with a mean of  $-11.8 \pm 3.2\text{‰}$ , suggesting a majority of agriculturally-derived N mixed with combustion-derived N (Boltersdorf and Werner, 2014). Additionally, the mean  $\delta^{15}\text{N}$  value for the GT transect was  $-11.8 \pm 2.4\text{‰}$ , for the JHMR transect was  $-10.7 \pm 4.5\text{‰}$ , and for the AP transect was  $-14.0 \pm 1.9\text{‰}$ . This suggests that JHMR and GT receive more N from combustion sources, likely because they are in more heavily visited areas closer to cities. Furthermore,  $\delta^{15}\text{N}$  values increased with lichen %N (Figure 4), which suggests that sites that receive high  $N_{dep}$  receive more deposition from combustion sources (Zhang et al., 2008). However, high %N and higher  $\delta^{15}\text{N}$  values for sites near JHMR which may drive this trend. Additionally, there could be separation occurring in the different chemical pathways that lead  $\text{NO}_x$  and  $\text{NH}_x$  to be deposited.  $\text{NO}_x$  is highly soluble and is deposited predominantly as wet deposition, while  $\text{NH}_x$  forms aerosols and a larger fraction is deposited in dry deposition (Behera et al., 2013; Zhang et al., 2012).

This could lead high elevation sites and other sites that receive more precipitation to have higher  $\delta^{15}\text{N}$  values due to high  $\text{NO}_x$  deposition.

At two elevation transects at JHMR and GT, %N increased with elevation ( $p \leq 0.001$ ), and at the third transect near AP, elevation did not affect lichen %N (Figure 5). At JHMR and GT,  $N_{dep}$  likely increases with elevation due to orographic effects, which cause higher elevation sites to receive more precipitation and more wet N deposition (Clow et al., 2015; Weathers et al., 2006). The differences in observed elevation patterns may be because the JHMR and GT transects have more direct point sources of N, whereas the AP transect receives mainly regional N. Due to this, in the AP area,  $N_{dep}$  would come mainly from the thermal decomposition of peroxyacetyl nitrate from long distance transport, as it sinks towards the Earth's surface or from deposition of  $\text{NH}_4$  aerosols, which are not driven by precipitation and are more constant across elevation (Behera et al., 2013; Zhang et al., 2012). On average, across the GYE, modelling suggests that roughly 40% of  $N_{dep}$  originates from livestock or fertilizer  $\text{NH}_3$ , 30% from mobile combustion sources, and that 50% of all  $N_{dep}$  sources are more than 400km away (Lee et al., 2016).

## Conclusions

Preliminary analyses suggest that further understanding how lichens incorporate N will be critical given the large amount of variation in %N and  $\delta^{15}\text{N}$  between lichens collected at the same site. Generally, N deposition was higher in western areas of the GYE, likely because predominant winds come from the west and major N sources are to the west. Additionally, if there are nearby sources of N,  $N_{dep}$  will likely increase with elevation as atmospheric N is wet deposited. Also, isotope ratios suggest that  $\text{NO}_x$  is an important source at sites with high N deposition, which could be due to these high N sites being near Jackson, influence from vehicle combustion, or differences in the chemical pathways that  $\text{NH}_x$  and  $\text{NO}_x$  are deposited by. However, patterns of  $N_{dep}$  across small spatial scales are highly variable and are influenced by topography and climate patterns.



**Figure 5.** Lichen thalli %N at three elevation transects in the Greater Yellowstone Ecosystem: A) Grand Targhee Ski Resort, B) Jackson Hole Mountain Resort, and C) Avalanche Peak.

## Future Work

Analysis of several hundred lichen samples collected at additional sites will help us understand the spatial distribution of N deposition in the GYE at larger scales. We also established ion exchange resin collectors at the 15 sites described in this report, which measure  $\text{NO}_3^-$  and  $\text{NH}_4^+$  ions deposited in rainwater (Fenn and Poth, 2004). These data combined with site characteristics (including: live or dead trees, slope aspect) and climate data will help us develop a clearer understanding of the relationship between N deposition and lichen thalli %N and  $\delta^{15}\text{N}$ .

## Acknowledgements

We thank the UW-NPS Research Station and the Greater Yellowstone Coordinating Committee for financial support, our collaborators Jill McMurray and Shannon Albeke for ideas and advice, Erik Oberg, John Klaptosky and Sue Consuelo-Murphy for assistance with research permits, David Mayer, Will Bowers, and Ryan Reynolds for help with fieldwork, Charmaine Delmatier for advice and help in the field, the UW Stable Isotope Facility for analyzing samples, and Harold Bergman, Michael Dillon and Bonnie Robinson for their support and hosting at the research station.

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